Sb-terminated InAs(001)-(2×4) and (2×8) studied using scanning tunneling microscopy and *ab initio* density functional theory

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ABSTRACT

We have studied the structure of MBE-grown InAs(001)-(2×4) surfaces exposed to low Sb_2 fluxes by scanning tunneling microscopy (STM) and *ab initio* density functional theory (DFT). Experimentally, we observe an Sb-terminated α2(2x4) phase over a wide range of temperatures (400-510 °C) for low Sb_2 flux (<0.1 ML/s), whereas temperature and As_2 flux must be carefully controlled to achieve the same As-terminated surface structure. At lower temperatures, we observe indications of an Sb-terminated (2x8) symmetry surface phase, and we report briefly on its proposed structure and stability, as well as its possible role in subsequent formation of the Sb-terminated (1x3) phase found at typical Sb_2 fluxes used during heterostructure growth.

INTRODUCTION

The nearly lattice-matched 6.1Å system of III-V semiconductor materials (InAs, GaSb, and AlSb) is being investigated for use in a wide variety of novel optoelectronic and high-speed quantum devices. All of these devices incorporate very thin layers, and as a consequence, the interfaces between the constituent materials can have a disproportionate impact on device performance. Recent studies by Nosho *et al.* [1] have highlighted the need for understanding surface reconstructions and their effect on interface formation. For instance, during typical InSb-like interface formation between an AlSb or GaSb layer on an InAs layer, the transition between the (2x4) surface of the InAs and the (1x3)-like surface that results after Sb exposure can lead to monolayer-height interface roughness unless the stoichiometry between the two phases is taken into account. Even during heteroepitaxy involving GaSb and AlSb, reconstruction transitions may lead to undesirable roughness at heterostructure interfaces.[2] Hence, knowledge of surface structure under varying conditions of temperature and flux can be a valuable aid to a device grower in devising interface preparation techniques for tailored interface roughness.

In this work, we seek to extend the studies of Nosho et al. into the low Sb flux regime to more fully explore the stable and metastable Sb-on-InAs phases, their structure, and implications for interface formation. We find that at the lowest Sb fluxes, an α2-(2×4) phase is present and stable on the surface up to 510°C. The desorption rate of Sb from this surface is exceedingly slow, and surfaces can be stabilized for long periods of time at lower temperatures < 480°C. As the substrate temperature is decreased, a (2×8) structure appears that apparently incorporates multiple layers of Sb and appears structurally identical to that found on GaAs. At higher Sb_2 fluxes and substrate temperatures, such as might be used for typical device growth, (F_{Sb_2}>1-2 ML/s) the surface transitions fully to a (1×3)-like phase, in agreement with previous observations. At low temperatures and intermediate fluxes, the surface is comprised of a mixture of (2x4) (2x8) and (1x3) structures. Given the progression of structures with

increasing Sb flux, and their arrangement on mixed-phase surfaces, we propose a simple mechanism for formation of the (1x3)-like surface during typical interface formation techniques used in the MBE growth of InAs/III-Sb structures.

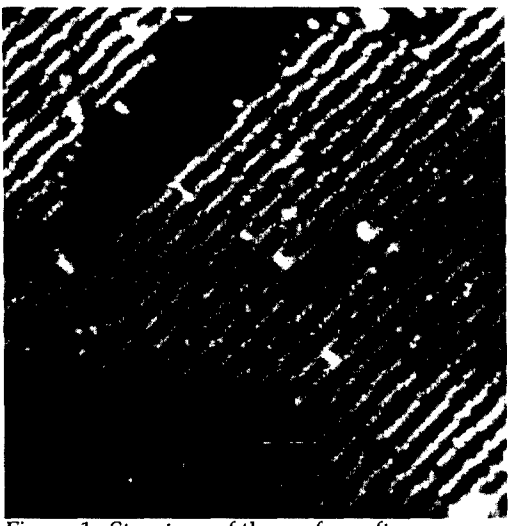

Figure 1. Structure of the surface after exposure to an Sb_2 flux of 0.1 ML/s at 470°C. Image is 50nmx50nm, filled states.

EXPERIMENT

The surfaces we report in this paper were prepared using a VG 80H MBE chamber, starting with unintentionally doped, 0.5 to 1 μm thick InAs buffer layers grown on nominally 0.05° miscut n-type (undoped) InAs(001) substrates at 470°C. As_2 flux rates were measured using the uptake method[3,4] on InAs. Sb_2 flux rates were estimated by the threshold flux needed to stabilize III-Sb growth. After various surface treatments, as described in the text, samples were quenched by simultaneously cutting the substrate heater power, shuttering all sources, rotating the sample towards the cryopanel, followed by rapid removal of the substrate from the chamber. The samples were then transferred *in vacuo* to a connected UHV chamber containing a full-wafer Omicron STM [5] where they are placed in contact with a cold metal block. After ~30 minutes, STM images can be acquired with little thermal drift. All filled- and empty-states STM images were acquired in constant current mode using bias voltages between 1.1 and 3.0 V and tunneling currents between 0.03 and 0.2 nA.

RESULTS AND DISCUSSION

In figure 1, we present the structure of the InAs surface at 470°C stabilized by an Sb_2 flux of ~0.1 ML/s. The surface prior to quench is apparently well ordered in the RHEED pattern, and this is reflected in the surface structure observed by STM. Interestingly, while the rows are well ordered in the 2x and x4 directions, there is disorder within each unit cell. From a high-resolution image of this structure, shown in figure 2, we observe that the structural units along the rows appear nearly identical to those observed on As-terminated InAs surfaces. In particular, the dominant reconstructed unit on the surface

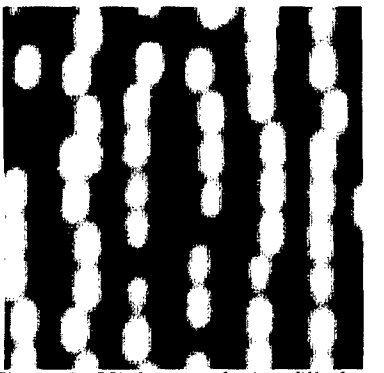

Figure 2. Higher resolution filled-states image of the α2(2x4) structure formed when exposing InAs(001) to Sb_2 flux.

appears to have the α2-(2×4) structure. Shown schematically in figure 3, the structure consists of a single Sb dimer atop a row of 3 In atoms, with a trench between each row. The In atoms at the edge of the row rebond with In atoms supporting the top As dimer. The trench dimer is also replaced with Sb. Theoretically, we find this structure to be the most stable relative to a number of other candidate structures[6].

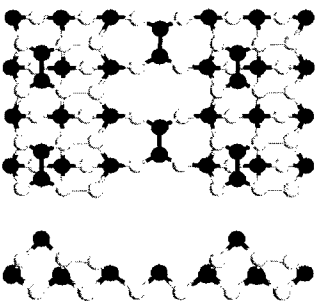

Fig. 3. Ball-and-stick model of the Sb-terminated α2-(2×4) structure. White balls represent In, grey balls represent As, and black balls represent Sb dimers.

While there are clear similarities in the structure of the unit cell between As- and Sb-terminated (2x4) surfaces, some differences emerge as well. For instance, the longer-range structure of the surface also changes dramatically. For comparison, in figure 4 we present an As-terminated α2-(2×4) surface, prepared by annealing an InAs(001) surface at 420°C for 20 minutes under an As_2 flux of 0.01 ML/s. Details concerning the surface phases and associated structures (e.g. the ad-structures clearly visible atop the rows in figure 3) can be found in a forthcoming publication [7]. Briefly, on As-terminated α2-(2×4) surfaces, the surface structure can by characterized by a large density of point and extended, island-like defects. Additionally, many defects in the dimer rows themselves are apparent, including vacancies and occasional β2-(2×4) reconstructed units. When the surface is converted to an Sb-terminated α2-(2×4), the surface appears much "cleaner" than the As-terminated surface. Additionally, we find an extremely small density of β2-(2×4) reconstructed units – one of the few observed β2 units can be seen in the middle left hand side of figure 2(a). This may possibly indicate that the β2-(2×4) structure is destabilized when the As terminated InAs(001) surface is exposed to Sb.

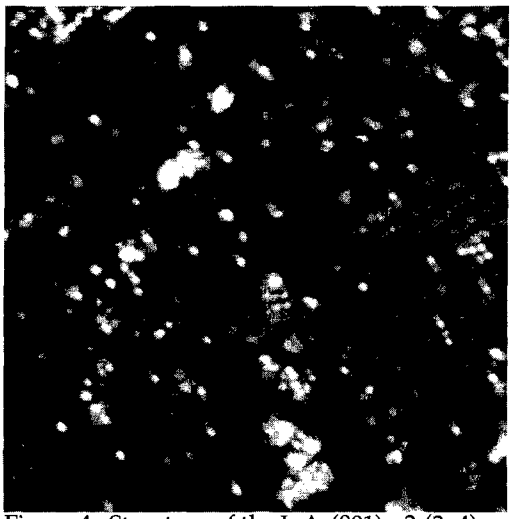

Figure 4. Structure of the InAs(001) α2-(2×4) surface after annealing under a flux of 0.01 ML/s at 420°C. Image is 50nmx50nm, filled states.

At higher Sb fluxes (>1 ML/s) at 470°C we find that the surface makes a transition to a (1×3) structure, as measured by RHEED, in agreement with prior observations.[1] Unlike the case of Sb on GaAs(001) [8] we find no evidence in RHEED that any intermediate phases form at these temperatures. However, we find at

Fig 5: Surface after cooling to 350°C under an Sb_2 flux of ~0.1 ML/sec. Filled states image, 50nmx50nm.

lower temperatures that an additional surface structure can form which has a (2x8) symmetry. Practically speaking, it is difficult to fully stabilize the surface at typical growth temperatures (420-470°C) in the (2×8) structure – only at lower temperatures does this appear to be possible.

In figure 5 we show an STM image of a surface that was cooled from 470°C to 350°C with an Sb_2 flux of ~0.3 ML/s. As judged from the symmetry of the surface as measured by RHEED, the α2-(2×4) structure is maintained until ~400°C, below which temperature ×8 diffraction spots gradually emerge until 350°C where they appear to saturate in intensity. This observation indicates that the (2×8) structure may not in fact be a thermodynamically stable structure, but is instead metastable. Another possibility is that entropic stabilization of energetically similar structures, like the α2-(2×4) and (1×3)-like, may squeeze out the (2×8) region of stability at higher temperatures.

From the arrangement of the α2-(2×4) and (2×8) structural units on the surface, we postulate a plausible structure and formation mechanism for the (2×8) phase. We suggest a simple structure for this phase, shown in figure 6. The structure is an extension of the α2-(2×4) phase, with the trench between neighboring α2-(2×4) units being filled with In, and the whole structure capped by a double layer of Sb. The structure fulfills electron-counting rules. Our structure is significantly different from the structure proposed by Whitman *et al.* [8] for Sb-on-GaAs-(2×8), which incorporated Sb_{In} antisite defects, no rebonded α2-like In atoms, and no trench dimer. Additionally, their structure is *asymmetric* with respect to the centerline of the unit cell, a quality that does not appear to be present in our images of this structure.

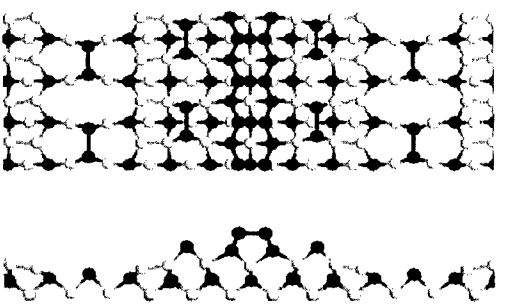

Figure 6. Proposed structure of the (2x8) surface phase. White balls represent In atoms, light grey represent As atoms, and dark balls represent Sb atoms and dimers.

We have tested the stability of both our proposed structure and that proposed by Whitman *et al.* using

density functional theory (DFT). The DFT calculations were carried out using the FHI98MD simulation package [9] within the local density approximation using norm-conserving pseudopotentials [10]. The electronic wave functions were expanded in a plane wave basis that was truncated at a cutoff energy of 12 Ry. The equivalent of 8x8 k-points in a 1x1 unit cell were used for k-point summation, generated using the scheme proposed by Monkhorst and Pack [11].

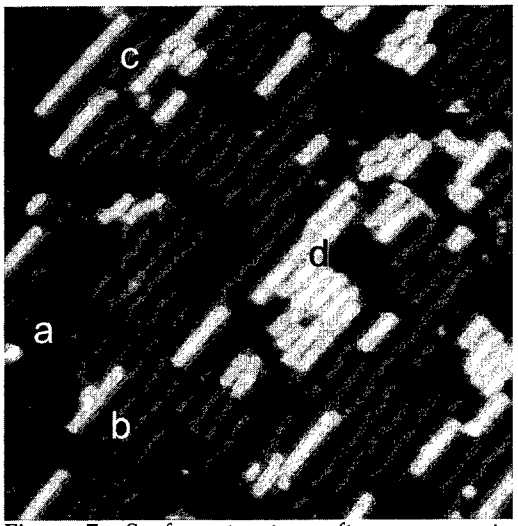

Figure 7. Surface structure after exposure to 10s Sb_2 at 350°C. All major surface phases are present: (a) disordered 1st layer, (b) α2-(2x4), (c) (2x8)-like structures and (d) 2nd layer (1x3). Image is filled states, 50nm x 50nm.

We find that our proposed structure is more stable. This is perhaps not surprising – our proposed structure incorporates only structural motifs from known stable phases. The edges and trench of the structure are identical to those found in α2-(2×4). The top of the structure, involving multiple layers of Sb, is qualitatively similar to the multilayer Sb reconstructions of the III-Sb's [12]. Additionally, while electron counting is fulfilled for both structures, Sb_{In} defects can involve a significantly higher energy cost.

At still higher Sb_2 fluxes, we begin to observe the formation of the (1×3)-like surface observed by Nosho *et al.* [1]. In figure 7, we show STM of a surface, stabilized initially at an Sb flux of ~0.1ML/s at 490°C, and then cooled to 350°C where the surface was exposed to a higher Sb flux (~0.5 ML/s) for 10 minutes. The resulting surface clearly has α2-(2×4), (2×8), and (1×3)-like structures present. There is an additional disordered structure, labeled "a" on the figure, which appears to be lower topologically than neighboring (2×4) structures, which may represent areas of the surface where In atoms have been removed for incorporation into neighboring (2×8) and (1×3)-like regions of the surface. As the In atoms in the α2-(2×4) structure are removed, the underlying As is exposed. Ideally these regions can form a quasi (1×3)-like structure with Sb capping the As exposed by the departing In. In this manner, these regions would evolve into (1×3)-like regions that are topologically 1 ML lower than nearby (1x3) "islands". Subsequent desorption of top Sb and buried As dimers, followed by re-capping with Sb dimers may allow these areas to eventually obtain the "ideal" multilayer Sb (1x3) or (4x3)-like surface structure.

Given the arrangement of these various structures on the mixed-phase surface in figure 7, we propose a simple mechanism for formation of the (1x3)-like surface. At typical growth temperatures, the desorption rate of As is much greater than Sb, leading to rapid conversion of the initial As-terminated (2x4) surface to an Sb-terminated α2-(2×4) phase. Once formed, there may be a high driving force for incorporation and

capping of In adatoms by adsorbing Sb-dimers between two α2 rows. Once a sufficient number of In atoms are capped between these two α2 rows, such an arrangement is nearly identical to the (2x8) structure shown figure 6. As shown in figure 7, there do not appear to be any neighboring (2x8) units on the surface. Therefore, we speculate that once two neighboring (2x8) units are formed, the top layer Sb might either be much more mobile, or have a higher desorption rate, enabling reorganization of this layer to form the (1x3) structure. Hence, although apparently not stable as a *surface phase* at higher temperatures, it is possible that (2×8)-like structures may mediate the transition from As-terminated InAs (2×4) surface to the (1×3)-like Sb-terminated surface during typical interface formation techniques used in the MBE growth of InAs/III-Sb structures.

CONCLUSIONS

At low Sb_2 fluxes, ~0.1 ML/s, we find that an α2-(2×4) phase is present and stable on the InAs(001) surface up to ~510°C. As the substrate temperature is decreased, a (2×8) structure appears. At low temperatures and intermediate fluxes, we observe a surface comprised of a mixture of (2x4) (2x8) and (1x3) structures. The arrangement of these structures on the surface, along with our proposed structure for the (2x8), leads us to propose a simple mechanism for formation of the (1x3)-like surface during typical interface formation techniques used in the MBE growth of InAs/III-Sb structures. By tailoring the starting surface for interface formation (for instance, instead of a (1x3), use the (2x8) or the α2) it may be possible to advantageously change the resulting distribution of group III and group V atoms at a heterostructure interface.

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